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High-Resolution Compton-Suppressed CZT and $LaCl_3$ Detectors for Fission Products Identification

Rahmat Aryaeinejad, John K. Hartwell, and Wade W. Scates

Abstract-- Room temperature semiconductor CdZnTe (CZT) detectors are currently limited to total detector volumes of 1-2 cm^3, which is dictated by the poor charge transport characteristics. Because of this size limitation, one of the problems in accurately determining isotope identification is the enormous background from Compton scattering events. Eliminating this background will not only increase the sensitivity and accuracy of measurements, but will also help to resolve peaks buried under the background and peaks in close vicinity of others. We are currently developing a fission products detection system based on the Compton-suppressed CZT and $LaCl_3$(Ce) detectors. In this application the detection system is required to operate in a high radiation field. Therefore, a small 10 mm × 10 mm × 5 mm CZT and ∅13 mm × 15 mm $LaCl_3$ detector are placed inside the center of a well-shielded ∅76 mm by 76 mm long NaI detector. So far, we have been able to successfully reduce the Compton background by a factor of 3.7 to 4.0 for a ^{137}Cs spectrum. In this work, we will discuss the performance of this detection system using both CZT and $LaCl_3$ detectors. The results are compared with MCNP calculations.

I. INTRODUCTION

Existing room temperature detectors such as CZT semiconductor detectors, although relatively inexpensive and capable of isotope identification, have limited detection efficiency due to their small volume (about 1-2 cm^3) [1]. This is because the larger detectors result in poor charge collection at room temperature, which in turn results in poor energy resolution. One way to increase the volume without sacrificing the energy resolution is by using an array of detectors and stacking them on top of each other [2]. A single and small CZT detector sometimes has difficulty accurately determining isotope identifications in an energy range below 1 MeV because of background from Compton scattering events. These background events decrease the sensitivity and accuracy of detector measurements, bury the weak gamma-ray peaks under the background, and mask those peaks that are in close vicinity to others. In a small CZT detector, the background above which gamma-ray peaks must be detected is a result of the mechanisms of gamma-ray interactions in matter, rather than the result of ambient background gamma rays reaching the detector. The two interaction mechanisms we are concerned with are photoelectric absorption and Compton scattering. Both of these interactions take place with atomic shell electrons of detector materials and result in either complete or partial transfer of the gamma-ray photon energy to electron energy. In Compton scattering interactions, the incoming photon transfers only a portion of its energy to the electron and is deflected through an angle θ with respect to its original direction. Since all angles of scattering are possible, the energy transferred to the electron can vary from zero to a large fraction of the gamma-ray energy. If a full energy photon undergoes a Compton scattering event in the sensitive volume of a radiation detector, and the scattered photon escapes, it results in a count in the broad continuum below the full energy peak. Compton scattering in the detector, especially in a small CZT detector, is the primary contributor to gamma-ray backgrounds. Since the scattered photons are always degraded in energy from the full energy photon, the continuum in multiple peak gamma-ray spectra increases rapidly at lower energies. This is why techniques must be implemented to decrease the contribution of Compton scattering within the detector. Also, the probability that a scattered photon will escape the detector volume and contribute to the spectral continuum is inversely proportional to the total detector volume. Large volume detectors, in addition to having better photopeak efficiency, generally have better ratios of photopeak-to-Compton (P/C) continuum counts. Therefore, one way of increasing the full energy peak and decreasing the Compton scattering events is by using a larger detector. However, at the present time, the maximum size in a CZT detector is limited by charge collection considerations to a volume of about 15 mm × 15 mm × 7.5 mm with an energy resolution of 2 to 2.5% at 662 keV.

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II. DETECTION SYSTEM

Active techniques to improve the P/C ratio of HPGe detectors have been successfully used in basic nuclear physics research and environmental applications over the last 35 years [3]-[7]. In these systems HPGe detectors are used as primary high-resolution detectors, and NaI or BGO scintillators are used as active Compton shields. Well-designed anti-Compton guards used with HPGe detectors can reduce the Compton continuum over the energy range of interest by factors of 5 to

10, depending on the size of both primary detector and the guard detector and also the type of guard detector. This can be accomplished with no impact on the detection of full energy photopeak. In these techniques the primary detector is placed inside an active anti-Compton shield used as a second veto detector. When a Compton scattering event occurs in the primary detector and the scattered photon escapes the crystal, the veto detector detects the scattered gamma ray and generates an output pulse that can be used in conjunction with an anti-coincidence circuit to block storage of the primary detector partial energy event. The veto detector need not be a high-resolution detector, but it must have high efficiency for the detection of scattered photons over the energy of interest. Thus, scintillation detectors such as BGO and NaI are commonly used. The BGO detector has higher density than the NaI detector and consequently more stopping power. However, it is heavier and more expensive. Ultimately, the reduction of background will definitely enhance the quality of the gamma-ray spectrum in the information-rich energy range below 1 MeV, which consequently increases the detection sensitivity.

P. Peerani et al. [8] have used a Ge Compton suppression gamma spectrometer for the detection of fission products at the trace levels in environmental samples. They were able to improve the detection limits for ^{137}Cs by a factor of about 3, and for the other nuclides, by a factor of 1.6-2.4. T.H. Prettyman, et al. has developed Compton-suppressed CZT detection systems for space and safeguards applications [9]. Their design placed the CdZnTe detector in a shallow well in a BGO veto detector, leaving the entire entrance face of the primary detector unguarded. This configuration achieved a Compton suppression factor of about 2.

Our application, on the other hand, is for online monitoring and identification of fission products produced in a reactor. This means that the detection system will be required to operate in a high radiation field. Therefore, small primary detectors with a small collimator and heavy passive shielding around the active anti-Compton guard detector are needed. Fig. 1 shows the detection system that we are developing for this specific application. It consists of a NaI anti-Compton guard detector and a small CZT detector. The NaI guard detector is ∅76 mm × 76 mm and was designed by us and manufactured by Saint-Gobain Corporation. It has two perpendicular ports: one on the side providing access for a primary detector and the second port in the front for admitting source gamma-rays. Both ports are 15.2 mm in diameter. The thickness of the NaI is 35.5 mm in the front and 40.6 mm in the back of the detector port. The % absorption for 1 MeV gamma-ray energy is 50% through the side, 55% through the back, and 64% though the back diagonally. The absorptions for the same design of a BGO anti-Compton guard are 78%, 83%, and 90%, respectively. The Tungsten collimator in the front of the NaI detector is 100 mm × 100 mm × 38 mm. It has a hole in the center to allow collimators of 3.2 mm, 6.4 mm or 12.8 mm to be inserted. A Teflon holder (see Fig. 1) in the front of the collimator centers the radioactive disc sources for test measurements. The source-to-detector distance is typically 100 mm.

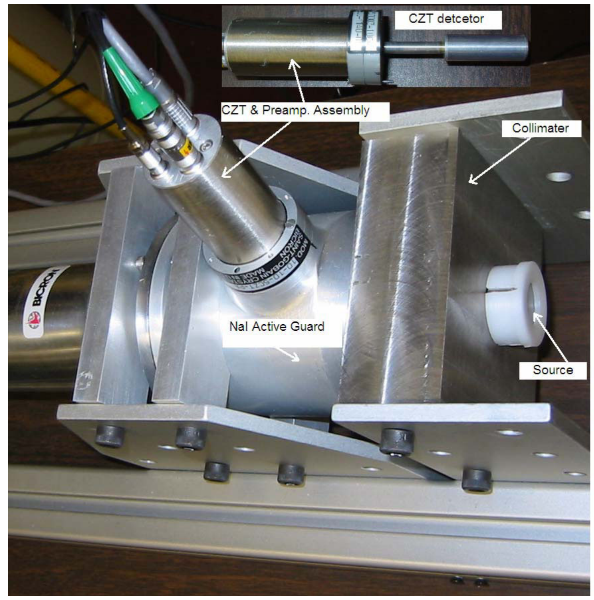

Fig. 1. A photograph of the Compton-suppressed CZT detection system for fission products identification.

Measurements were made with two primary detectors; a 10 mm × 10 mm × 5 mm CZT detector, and a ∅13 mm × 15 mm $LaCl_3$(Ce) scintillation detector using a 6.4 mm collimator. In the design of anti-Compton detection systems, it is extremely important to minimize the amount of inactive material between the primary spectrometry detector and the guard detector since scattered photons that are stopped in "dead" layers cannot be vetoed. Therefore the CZT detector shown on the top of Fig. 1 was designed with this principle in mind. We mounted the CZT detector on a thin circuit board and placed it inside an aluminum cylinder of ∅13 mm × 30 mm. The detector was oriented so that photons entering through a designated side of the Al cap entered the detector through the cathode. Signal cables were sent through a small Aluminum tube to the preamplifier outside the anti-Compton shield. This design minimizes the amount of materials around the primary detector. However, it degrades the detector resolution due to added cable capacitance between the detector and the preamplifier located outside. The CZT detector resolution was measured to be about 3.9% for ^{137}Cs peak. The $LaCl_3$(Ce) detector was also designed to fit inside the same guard with a combined detector and photomultiplier dimension of was ∅15 mm by 105 mm long, again with the preamplifier located outside the guard detector. Its resolution was 5.9% at 662 keV.

III. MEASUREMENTS AND SUMMARY OF THE RESULTS

A simple electronics setup shown in Fig. 2 was used for all measurements. Output of the primary detector was sent to the Tennelec linear amplifier (Model TC-244) and then to the multichannel analyzer (ORTEC Nomad MCA). The Guard NaI output was fed into the timing filter amplifier (ORTEC TFA- 474). The Phillips discriminator (Model 710) was used to set the threshold and then send it to a Phillips 794 gate and delay generator to provide the gate to the MCA and acquire

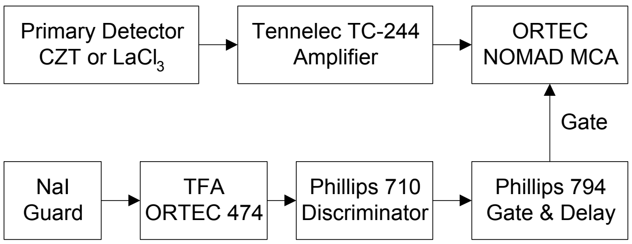

Fig. 2. Electronics diagram for active anti-Compton detection system.

suppressed and rejected spectra in anti-coincidence and coincidence modes, respectively.

Fig. 3 shows the spectra taken with this detection system using a ^{137}Cs source (662 keV) and the CZT detector (top spectra). These spectra are for unsuppressed data (black), Compton events that are rejected (red), and Compton suppressed data (Blue). The unsuppressed spectrum has a P/C ratio of 1.48 and the suppressed has a P/C ratio of 5.90. This is an improvement by a factor of 4.0. This ratio is defined by the number of counts in the maximum photopeak channel to the average continuum from 358 to 382 keV for the 662 keV gamma rays emitted from a ^{137}Cs source [10]. It is important to note that the rejection of Compton events has no impact on

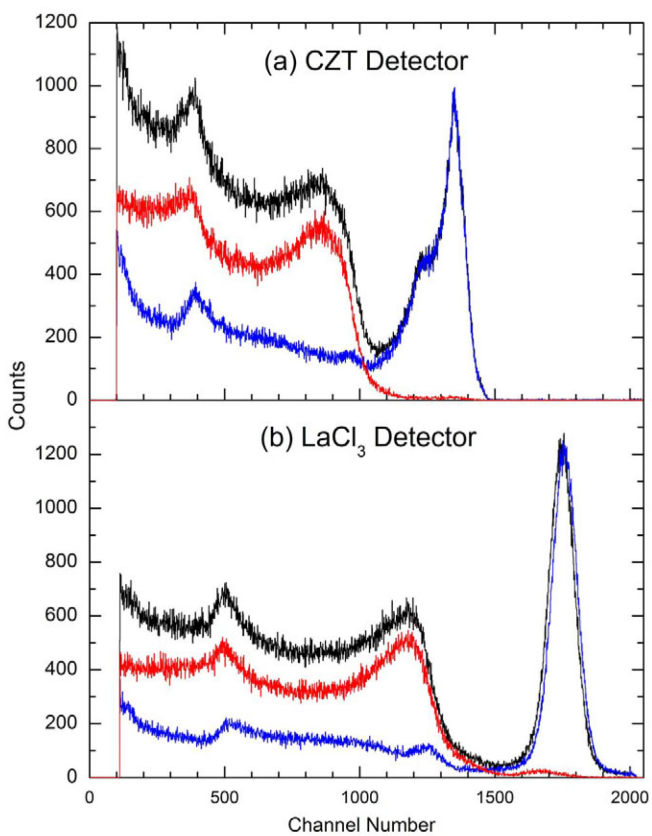

Fig. 3. A comparison of the spectra of ^{137}Cs source acquired with two Compton suppression detection systems, top spectra (a) are taken with the CZT detector and bottom spectra (b) are taken with the $LaCl_3$ detector.

the detection of a full energy photopeak at 662 keV. In other words, there is no photopeak present in the rejected Compton spectrum (red spectrum). Also, the Compton rejection is better at higher energies than at lower energies. This is primarily due to low-energy Compton events being stopped by dead materials between the CZT detector and the active NaI guard detector. Clearly, any reduction of the background under the peaks, or narrowing the peak widths translates into improved detection sensitivity.

The Compton suppression spectra for the $LaCl_3$ detector are shown at the bottom of Fig. 3. Again, the spectra are unsuppressed (black), Rejected (red), and suppressed (blue). Because the thickness of $LaCl_3$ is 13 mm as compared to CZT detector of 5 mm, one would basically expect to see a better peak to Compton ratio for the $LaCl_3$ detector. In fact the P/C ratios for this detector are 2.59 for the unsuppressed spectrum and 9.50 for the suppressed spectrum. This is an improvement in peak to Compton ratio by a factor of 3.7.

Table 1. Summary of the results for two primary detectors, CZT and LaCl3, for the 662 keV gamma-rays emitted from a ^{137}Cs source.

Detector	Type	P / C	Factor[a]	P_A / T_A	Factor[a]
CZT	Unsupp[b]	1.48	4.00	0.18	2.30
	Supp[b]	5.90		0.41	
$LaCl_3$	Unsupp	2.59	3.70	0.19	2.32
	Supp	9.50		0.44	

[a] Improvement factor defined as a ratio of Supp To Unsupp data.

[b] Unsupp and Supp are spectra taken without and with Compton suppression, respectively.

Table 1 shows the summary of the results from our measurements. The P_A/T_A ratio is the peak area divided by the total counts in the spectrum above the threshold, which was set just above the ^{137}Ba X-rays. There are only small differences in these ratios between the two detectors, and therefore the improvement factor in the last column is basically the same (2.30 versus 2.32).

We performed the same measurements with both primary detectors using a smaller collimator of 3.2 mm in diameter. The results showed no significant differences except much lower statistics due to the smaller collimator size.

IV. Comparisons with Monte Carlo Calculations

Measurements were used to benchmark Monte Carlo calculations of the expected performance of active anti-Compton systems. The geometry of the suppression scheme was modeled as close as possible using the MCNPX transport code version 2.5.e. Using this transport code the pulse height spectra for both the $LaCl_3$ and the CZT detectors were calculated with and without suppression from the NaI Compton shield. Fig. 3 shows the results of calculations for the $LaCl_3$ primary detector compared with the experimental spectra. It is apparent that the model does not reproduce the spectra very well especially in the lower energy region. This is believed due to a lack of detailed information regarding the dead layers in both primary detector and anti-Compton of suppressed spectra, the calculated data are about one unit lower than their experimental counterpart. This is another indication of not including enough dead materials in Monte Carlo simulations. Despite of all these discrepancies, the improvement factors which are defined as the ratios of suppressed P/C over the unsuppressed P/C are amazingly close to measured value especially for the 10 keV thresholds.

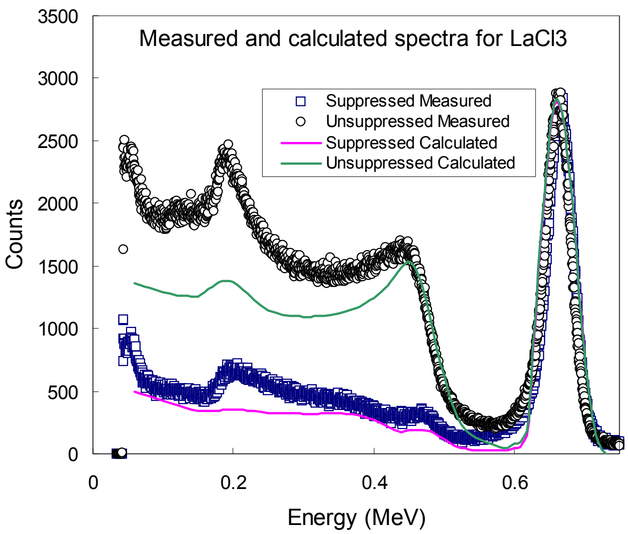

Fig. 4. A comparison of the spectra of ^{137}Cs source acquired with two Compton suppression detection systems, top spectra (a) are taken with the CZT detector and bottom spectra (b) are taken with the $LaCl_3$ detector.

shield. Therefore, some approximations were made about the amount of dead layers. These dead layers primarily include the detector housing, and the aluminum lining of the cavity in talking with the manufacturer. The back scatter peaks in unsuppressed and suppressed spectra are not predicted well in the calculations. As the thickness of the dead layer increases the backscatter peak becomes more pronounced. Two threshold levels of 10 and 20 keV were used for the NaI guard detector in the Monte Carlo calculations. The summary of the results are shown in Table 2 together with the experimental values.

Table 2. Comparisons between experimental data and Monte Carlo calculations for CZT and $LaCl_3$ detector using ^{137}Cs 662 keV gamma-ray.

Detector	Type	P / C			Factor[a]		
		Exp.	MCNP		Exp.	MCNP	
			10[b] keV	20[b] keV		10[b] keV	20[b] keV
CZT	Unsupp[c]	1.48	1.2	1.4	4.00	4.14	3.42
	Supp[c]	5.90	4.97	4.8			
$LaCl_3$	Unsupp	2.59	2.3	2.3	3.70	3.74	3.67
	Supp	9.50	8.6	8.45			

[a] Improvement factor defined as a ratio of Supp To Unsupp data.
[b] Two threshold settings of 10 and 20 keV were used in MCNP calculations for the active NAI guard detector.
[c] Unsupp and Supp are spectra taken without and with Compton suppression, respectively.

The calculated P/C ratios are always smaller (about 10%) than the experimental data for unsuppressed and suppressed spectra and for both the CZT and $LaCl_3$ detector. In the case

V. Conclusions

We have investigated the feasibility of using Compton-suppressed detection systems for monitoring and identifying fission products in a high radiation field produced in a reactor. Small room temperature semiconductor CZT and scintillator $Lacl_3$ detector were used in conjunction with an active NaI guard detector. Using this detection technique reduced the Compton scattered background in the information-rich energy range below 1 MeV. The results show that for the CZT detector a P/C ratios of 1.48 and 5.9 for the unsuppressed and suppressed spectra which is an improvement factor of 4. In the case of the $LaCl_3$(Ce) detector the P/C of 2.59 and 9.5 were obtained for the unsuppressed and suppressed spectra, respectively (a factor of 3.7 improvement). Clearly, any reduction of the background under the peaks, or narrowing the peak widths translates into improved detection sensitivity, which results in faster and better radiation detection and isotope identification.

Monte Carlo calculations indicate that the calculated P/C ratios are about 10% lower than experimental values for unsuppressed and suppressed spectra. This is the case for both the CZT and $LaCl_3$ detector. However, the Improvement factors are very close to the experimental data especially if we use 10 keV thresholds for the NaI guard detector.

In the future we plan to address electronic issues such as timing systems for effective veto that will serve as a baseline and either use a BGO anti-Compton shield or a thicker NaI guard detector. We strongly believe that with a proper and sophisticated guard design, we can achieve Compton reduction factors of 8 or better. The key technical challenges we anticipate will be to optimize the active guard, reduce the amount of dead layers in both the primary and guard detector. This will ultimately improve the sensitivity of the detection system so it becomes more suitable for monitoring and identifying fission products.

VI. Acknowledgments

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References

[1] T.H. Prettyman, M.K. Smith, and S.A. Soldner, "Design and characterization of cylindrical CdZnTe detectors with coplanar grids," *Proc. SPIE,* vol. 3768, pp. 339–347, 1999.

[2] T.H. Prettyman, W.C. Feldman, K.R. Fuller, S.A. Storms, S.A. Soldner, C. Szeles, F.P. Ameduri, D.J. Lawrence, M.C. Browne, and C.E. Moss,

"CdZnTe Gamma Ray Spectrometer for Orbital Planetary Missions," *IEEE Trans. Nucl. Sci.,* vol. 49, no. 4, pp. 1881-1886, August 2002.

[3] P.J. Twin, P.J. Nolan, R. Aryaeinejad, D.J.G. Love, A.H. Nelson, And R. Kirwan, "TESSA : A Multi-Detector Gamma Ray Array Designed to Study High Spin States," *Nucl. Phys.*, A409, pp. 343c-351c 1983.

[4] P.J. Nolan, D.W. Gifford, and P.J. Twin, "The Performance of a Bismuth Germanate Escape Suppression Spectrometer," *Nucl. Instr. and Meth*., A236, pp. 95-99, 1985.

[5] C.W. Beausang, S.A. Forbes, P. Fallon, P.J. Nolan, P.J. Twin, J.N. Mo, J.C. Lisle, M.A. Bentley, J.C. Simpson, F.A. Beck, D. Curien, G. deFrance, G. Duchêne, D. Popescu, "Measurements on prototype Ge and BGO detectors for the Eurogam array," *Nucl. Instr. and Meth*., A313, pp. 37-49, 1992.

[6] M.P.Carpenter, T.L. Khoo, I. Ahmad, R.V.F Janssens, T. Lauritsen, G.A. Annan, A.M. Baxter, M.E. Bleich, S. Harfenist, E.F. Moore, M. Torelli, D.W. Visser, "Test and performance of a BGO Compton-suppression shield for GAMMASPHERE," *Nucl. Instr. and Meth*., A353, pp. 234-238, 1994.

[7] E. Mauerhofer, U. Tharun, H.O. Denschlag, R. Schmidt, J.V. Kratz, "A Compton suppression spectrometer for neutron activation analysis," *Nucl. Instr. and Meth*., A371, pp. 665-471, 1996.

[8] P. Peerani1, P. Carbol, E. Hrnecek, M. Betti, "Assessment of a Compton-event suppression gamma spectrometer for the detection of fission products at trace levels," *Nucl. Instr. and Meth*., A482, pp. 42-50, 2002.

[9] T.H. Prettyman, F.P. Ameduri, R. Arlt, J. Audia, G.L. Carlone, E.E. Chumbley, W.C. Feldman, K.R. Fuller, K.D. Ianakiev, V. Ivanov, K.R. Moore, J. Stein, and S.A. Storms, "Development of CdZnTe Detectors for Safeguards," *Proc. 42nd INMM Annual Conference,* Indian Wells, CA July 2001.

[10] ANSI/IEEE Standard 325-1986, "Test Procedures for Germanium Gamma Ray Detectors", 1986.